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Physical Controls on Total and Methylmercury Concentrations in Streams and Lakes of the Northeastern USA

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Abstract. The physical factors controlling total mercury (HgT) and methylmercury (MeHg) concentrations in lakes and streams of northeastern USA were assessed in a regional data set containing 693 HgT and 385 corresponding MeHg concentrations in surface waters. Multiple regression models using watershed characteristics and climatic variables explained 38% or less of the variance in HgT and MeHg. Land cover percentages and soil permeability generally provided modest predictive power. Percent wetlands alone explained 19% of the variance in MeHg in streams at low-flow, and it was the only significant ($p < 0.02$) predictor for MeHg in lakes, albeit explaining only 7% of the variance. When stream discharge was added as a variable it became the dominant predictor for HgT in streams, improving the model r^2 from 0.19 to 0.38. Stream discharge improved the MeHg model more modestly, from r^2 of 0.25 to 0.33. Methylation efficiency (MeHg/HgT) was modeled well (r^2 of 0.78) when a seasonal term was incorporated (sine wave with annual period). Physical models explained 18% of the variance in fish Hg concentrations in 134 lakes and 55% in 20 reservoirs. Our results highlight the important role of seasonality and short-term hydrologic changes to the delivery of Hg to water bodies.

Keywords: total mercury; methylmercury; SPARROW; land use; high flow events

Introduction

Many factors affect the distribution of total mercury (HgT) and methylmercury (MeHg) in surface waters. Some factors, such as basin size, land use/land cover, geology, soil properties, acid/base status, average climatic variables, and annual Hg deposition, vary primarily in space. Others, such as stream discharge, redox conditions, and other water chemistry indices, also vary in space but vary significantly in time, exhibiting both seasonal and short-term variations. In this paper, we conduct an empirical analysis of the Northeast Research Consortium (NERC) Hg data set (Evers and Clair, this issue) to assess the spatial and temporal factors that control HgT and MeHg concentrations in water and in fish muscle tissue in the streams and lakes of the northeastern USA.

Forests enhance Hg deposition as trees effectively scavenge Hg vapor through leaf stomata (Rea et al., 2002; Ericksen et al., 2003). Thus streams draining forested landscapes, particularly high-latitude and/or wetland-dominated systems that generate dissolved organic carbon (DOC)

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tend to have elevated Hg in waters, sediments, and biota (Mierle and Ingram, 1991; Driscoll et al., 1994; Kamman et al., 2004). High-DOC landscapes where reducing conditions occur in the presence of sulfate favor MeHg production and export (St. Louis et al., 1994; Branfireun et al., 1996). Thus forested uplands may be both a source of MeHg and a source of HgT that may subsequently be methylated in low-lying wetlands.

Surface-water HgT and MeHg concentrations are affected by watershed characteristics and land use classes, particularly by the amount, type, and distribution of wetlands (Hurley et al., 1995; Watras et al., 1995; St. Louis et al., 1996; Babiarz et al., 1998; Balogh et al., 1998; Grigal, 2002). Superimposed on these landscape controls are seasonal and event dynamics that tend to increase concentrations and fluxes of HgT and MeHg through increased water flux, sediment movement, and DOC increases (Babiarz et al., 1998; Hurley et al., 1998; Quémerais et al., 1999; Schwesig and Matzner, 2001). In particular, hydrologic events such as snowmelt and summer storms can be very important to the flux of Hg (Bishop et al., 1995b; Scherbatskoy et al., 1998; Allan et al., 2001; Shanley et al., 2002). Where both Hg and MeHg have been measured during high-flow events, HgT generally increases while MeHg dilutes with increasing flow (Bishop et al., 1995b; Lee et al., 2000; Munthe and Hultberg, 2004) in contrast to the coupled behavior of HgT and MeHg typically observed at base flow (Lee et al., 2000; Dennis et al., this issue).

Watershed disturbance is an additional factor that can mobilize HgT and MeHg in watersheds. Forest harvesting, road construction, or fire can lead to prolonged elevated concentrations of Hg and especially MeHg (Garcia and Carignan, 2000; Grigal, 2002; Porvari et al., 2003; Munthe and Hultberg, 2004). The effect of prolonged disturbance, e.g. land clearing in New England in the 19th century, is evident in elevated Hg concentrations in lake sediment profiles (Kamman and Engstrom, 2002). Finally, the short-term "disturbance" of fluctuating water levels in reservoirs results in elevated MeHg production due to redox cycling, resulting in increased MeHg concentration during reservoir refilling (Kelly et al., 1997; Lucotte et al., 1999; St. Louis et al., 2004).

The primary objective of this paper is to identify the watershed characteristics, climatic variables, and hydrologic conditions that control HgT and MeHg concentrations in the rivers and lakes in the NERC study areas of northeastern North America (Evers and Clair, this issue). A secondary objective is to evaluate the relation between these factors and Hg concentrations in fish. Dennis et al. (this issue) demonstrated a strong association between HgT and DOC within this data set, and noted that poor drainage conditions tend to increase HgT concentrations at base flow conditions. This paper builds on these results by evaluating a broader array of physical factors and hydrologic conditions, including the seasonality of controls on HgT and MeHg.

Methods

The NERC data set for stream and lake samples covered northeastern New York, New England and eastern Canada and was the same as that analyzed by Dennis et al. (this issue), except that the Adirondack and Canadian studies were excluded because the high-resolution physical and climatic parameters were not available for these areas. Also, (Dennis et al., 2005) excluded high-flow cases from their analysis but we retained those cases here. The resulting data set contained 693 cases, including 219 lake and 474 stream cases from 6 studies in the USA (Table 1). Each individual analysis was retained, so that the resulting data set includes multiple samples from some sites. These sites thus had a disproportionately high weight in the empirical models for their static properties (e.g. land cover percentages), but the trade-off was that time-varying parameters (stream discharge, seasonality) were well-represented by repeated sampling at some sites. Each sample had at minimum an analysis of total HgT. Of these 693 samples, 383 had analyses of MeHg. All Hg analyses were on unfiltered samples. For the few cases reporting both filtered and unfiltered analyses, the unfiltered values were used for consistency. There were too few filtered values to construct empirical models.

For each sampling site, we derived land use class percentages based on 42,000 stream reach watersheds from the SPARROW (SPAtially Referenced Regressions On Watershed attributes)

Table 1. Study project data sets compiled for this analysis and number of samples from each

Study name	Study region	Lake HgT	Lake MeHg	Stream HgT	Stream MeHg	Key references
NH/VT REMAP	NH/VT	202	202			Kamman et al. (2003)
Maine Rivers	Maine	2		102		Peckenham et al. (2003)
USGS Coastal Basins	RI to mid-Maine coast			74	74	Chalmers and Krabbenhoft (2001)
USGS Lake Champlain	Champlain basin: VT, NY, QUE	15	15	79	79	Shanley and Chalmers (2002)
USGS Sleepers River	Northeast Vermont			116	24	Shanley et al. (2002)
Nettle Brook	Northwest Vermont			77		Scherbatskoy et al. (1998)

database for New England (Moore et al., 2004). Each sampling location was matched to the pre-parameterized stream reach that contained it. Locations for which the match was poor (e.g. sampling points off stream) were not assigned SPARROW parameter values. The SPARROW database provided watershed land cover percentages (grouped to urban, agricultural, forested, wetlands, and open water classes to total 100%); mean annual temperature, precipitation, and runoff; mean soil permeability; and population density for the study watersheds. In addition to the SPARROW parameters, instantaneous stream discharge (if recorded) and a seasonal term (represented by sine and cosine waves with one-year period) were included in the empirical models (Table 2). No more than three of the five land use classes were included in any single regression model to avoid parameter covariance in a closed

Table 2. Physical parameters used as predictors in linear regression models

Parameter	Source
Watershed area	SPARROW[a]
Soil permeability	SPARROW
Mean annual precipitation	SPARROW
Mean annual temperature	SPARROW
Mean annual runoff	SPARROW
Population density	SPARROW
Percent agriculture	SPARROW
Percent forest	SPARROW
Percent urban	SPARROW
Percent wetland	SPARROW
Percent open water	SPARROW
Stream discharge per unit area	investigator
sine of day of year in radians	calculated from sample date
cosine of day of year in radians	calculated from sample date

[a]Moore et al. (2004).

data array. The NH/VT lake data set included standardized fish Hg concentrations. Additional lakes and reservoirs that did not have aqueous Hg were also included in the fish Hg analysis for a total of 134 lakes and 20 reservoirs.

For the aqueous Hg analyses, the data were grouped in three main categories: all lake samples, all stream samples, and a low-flow subset of stream samples. All stream discharges were normalized to basin area to obtain flow units of mm h^{-1}. Based on the distribution of flows we arbitrarily defined low flow as less than 0.08 mm h^{-1}. Cases without flow values known to be collected under base flow conditions were included in the low-flow set. Low-flow samples from the Maine rivers study were selected based on its low, medium, and high-flow designations (Peckenham et al., 2003). The resulting low-flow stream category contained 188 cases. Multiple linear regressions were performed for HgT and MeHg for each of the three categories, and for fish tissue mercury in each of the two categories (lakes and reservoirs) mentioned above. Stream discharge was removed as a variable in the low-flow streams category. Given the incomplete data matrix (e.g. missing stream discharges or SPARROW mismatch sites), adding or deleting a predictor variable from a regression could sometimes change the number of cases, thereby affecting the regression results. We qualified our interpretations accordingly.

Mercury deposition patterns

Mercury deposition generally decreases from SW to NE across the study region. Superimposed on this general trend are "hotspots" of enhanced deposition, which include high elevation areas, the coastal population corridor, and points downwind of discrete emission sources (Miller et al., this issue). Differences in Hg deposition and storage across the region may explain some of the differences in aqueous Hg concentrations that cannot be explained by watershed characteristics. However, Dennis et al. (this issue) found little correspondence of the geographical distribution of HgT and MeHg concentrations in surface waters to the regional Hg deposition gradient, except for an apparent association of high Hg concentrations with localized emission sources. Nonetheless, the tendency for watershed retention of Hg implies that areas of high depositon have potentially large stores of legacy Hg that may be released to receiving waters. Unfortunately, site-specific Hg deposition estimates were not available for this analysis.

Overview of aqueous Hg concentrations

The distribution of HgT concentrations was surprisingly similar in streams and lakes (Fig. 1). Total Hg concentrations ranged from 0.09 to 80 ng l^{-1} in streams and 0.22 to 35 ng l^{-1} in lakes. The median HgT concentration was 2.15 ng l^{-1} for streams and 1.90 ng l^{-1} for lakes, and interquartile ranges were also similar. In contrast to this similarity for HgT, the distribution of MeHg shifted to higher concentrations in lakes relative to streams. MeHg concentrations ranged from < 0.04

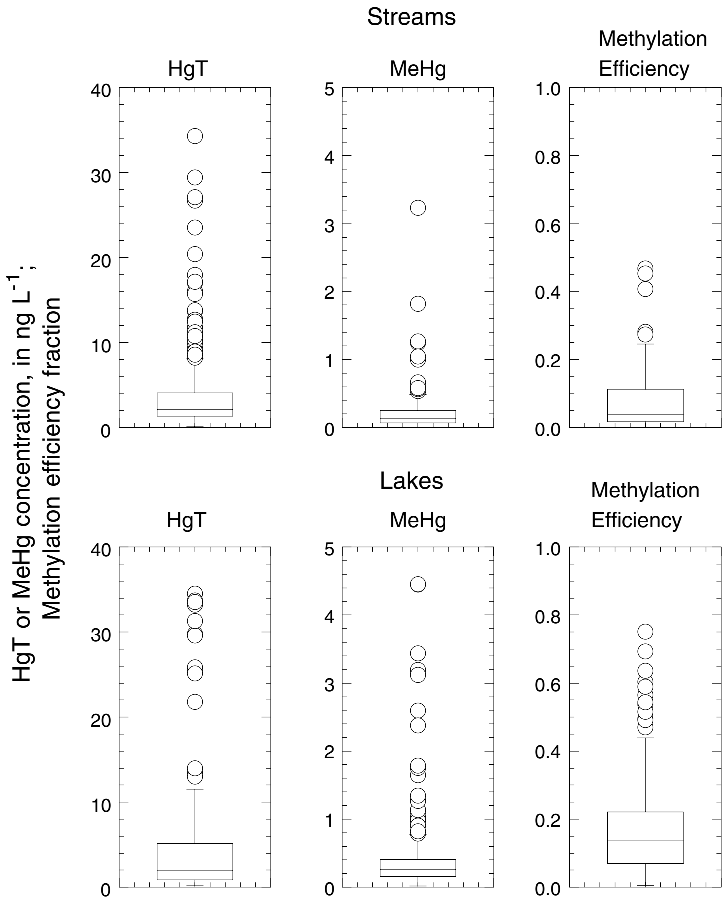

Figure 1. Boxplots of total mercury (HgT), methylmercury (MeHg), and methylation efficiency (MeHg/HgT) for streams and lakes in the data set. In each panel, the top and bottom of the wide box depicts the 25th and 75th percentiles of the data and the line within the box is the median. Tick marks above and below the box are the 10th and 90th percentiles, and circles are the individual values beyond these percentiles. In the HgT plot for streams (upper left), one outlier of 79.7 ng l^{-1} is not shown.

to 4.5 ng l^{-1} in lakes and <0.04 to 3.2 ng l^{-1} in streams. Median MeHg concentrations were 0.26 ng l^{-1} for lakes and 0.13 ng l^{-1} for streams. Accordingly, the median methylation efficiency in lakes was 13.9% compared to 3.9% for streams. Bear in mind that these methylation efficiencies are probably biased high due to the prevalence of summer sampling, especially for lakes.

Mercury in streams

Total mercury

For streams at low flow, each individual land cover class (in independent single regressions) predicted no more than about 10% of the variance in total Hg concentration. Percent forest was inversely correlated with HgT and explained 10% of its variability. This inverse relation may reflect the ability of the forest floor to retain Hg, despite the enhanced Hg deposition to the forest canopy (Rea et al., 2002) and despite enhanced DOC leaching from the forest landscape relative to other landscape types. The best multiple regression model using all static predictors explained 19% of the variance in streamwater HgT (Table 3), and percent forest was the most significant predictor. HgT concentration was inversely correlated to soil permeability, consistent with the notion that poorly drained soils promote near-surface runoff that flushes Hg from the forest floor. The negative

Table 3. Results of multiple linear regression models for total mercury (HgT) and methylmercury (MeHg) concentrations in north-eastern USA in three categories

Category	Species	*n*	Model r^2	Predictor	*p*-value
Low-flow streams	HgT	188	0.19	*Percent forest*	< 0.0001
				Soil permeability	0.0002
				Percent agriculture	0.004
				Annual precipitation	0.04
				Annual runoff	0.06
				Percent water	0.09
	MeHg	90	0.25	Percent wetland	< 0.0001
				Percent forest	0.013
				Soil permeability	0.03
All streams	HgT	254	0.38[a]	Stream discharge	< 0.0001
				Annual precipitation	< 0.0001
				Percent urban	0.0005
				Percent forest	0.002
				Annual runoff	0.011
	MeHg	79	0.33	Percent agriculture	0.007
				Stream discharge	0.011
				Population density	0.019
				Percent urban	0.06
				Annual precipitation	0.11
				Percent wetland	0.14
	Meff[b]	79	0.78	*Annual sine wave*	< 0.0001
				Annual runoff	0.004
				Percent water	0.013
				Percent urban	0.018
				Stream discharge	0.08
				Percent wetland	0.09
Lakes	HgT	204	no model		
	MeHg	202	0.09	Percent wetland	< 0.0001
				Percent forest	0.023
				Percent urban	0.07

n = number of cases in model; Predictors accepted in models only if $p < 0.25$. Predictors in italics have negative correlation.
[a] model r^2 decreases to 0.34 if one high HgT outlier is removed..
[b] Meff = methylation efficiency = MeHg/HgT.

effect of soil permeability parallels the findings of Krabbenhoft et al. (2004) and Dennis et al. (this issue). The latter authors calculated a drainage index for each site based on topography, and found that poor drainage promoted higher HgT concentrations in surface waters.

For stream analysis without the low-flow restriction, stream discharge was added as a predictor variable and 254 total cases were included. Some of the cases in the low-flow data set had no associated flow values (they were simply designated or assumed to be low-flow) so were excluded from this all-streams analysis. Therefore, the data sets overlap only partially and results cannot be compared directly. Nonetheless, stream discharge alone (single regression) explained 22% of the variance (28% for a log-log regression) in streamwater HgT (Fig. 2). This relation was strongly affected by a single high-Hg value, without which the variance explained was reduced to 12%. A full model with stream discharge and several physical and climatic parameters explained 38% of the variance in streamwater HgT. The variance explained fell to 34% without the single high-Hg value. The three leading predictors were stream discharge, annual precipitation, and percent urban (Table 3). For the Maine rivers study with its qualitative flow class designations, median HgT concentrations were 1.26 ng l^{-1} for low flow, 1.47 ng l^{-1} for medium flow, and 2.52 ng l^{-1} for high flow, consistent with a positive effect of stream discharge on HgT concentrations.

Methylmercury

A single regression of stream MeHg concentrations with percent wetlands explained 19% of the variation (24% for a semi-log regression) in MeHg in the low-flow samples (Fig. 3). Two outliers

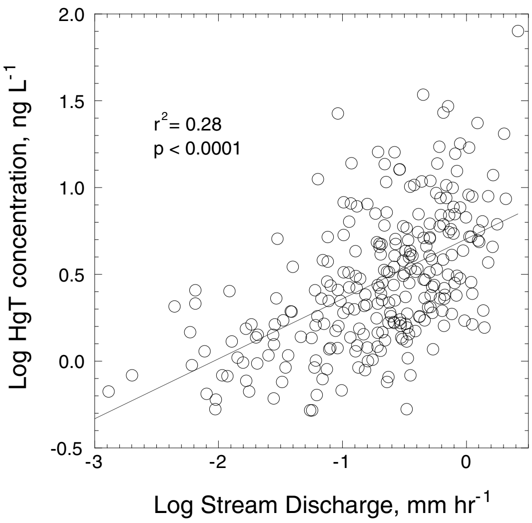

Figure 2. Log of total mercury concentration versus log of stream discharge for all stream cases with discharge values

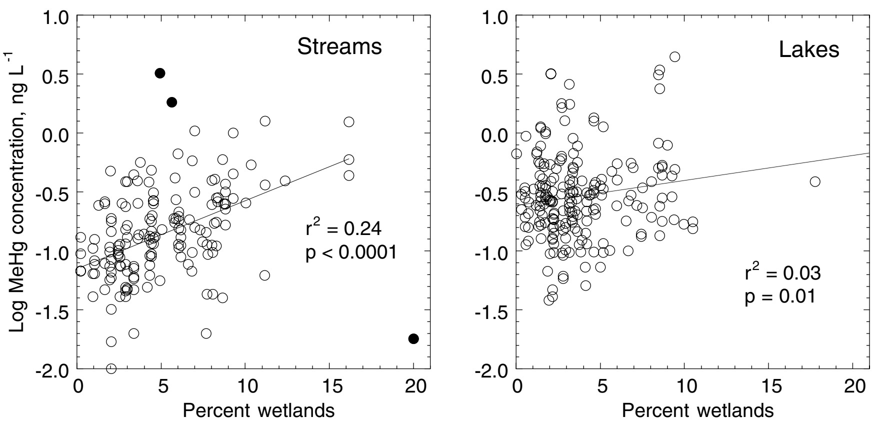

Figure 3. Log of methylmercury concentration versus percent wetlands for streams (left) and lakes (right). Solid circles in stream plot are outliers not included in the regression line shown for the plot. The two high-MeHg outliers (upper left) also were not included in the regression models, as discussed in text. The outlier at lower right was included in the models. This point represents a small watershed with high percent wetlands, but the wetlands are not hydrologically connected to the stream.

were removed from the regression but are included in Fig. 3 – these samples were collected near Boston and had very high MeHg concentrations but were near the mid-range of wetland percentage. We have no reason to question the MeHg concentrations but we suspect that a factor not captured by our model parameters, such as a localized point source, was responsible. The best full low-flow model for MeHg with the SPARROW output parameters explained 25% of the variance, and the full model including stream discharge explained 33% of the variance (Table 3), although stream discharge was second in importance to percent agriculture as a predictor. The positive effect of percent agriculture on MeHg concentrations is counterintuitive.

We had expected that MeHg concentrations in streams would display a stronger relation to percent wetlands. However, Grigal (2002) concluded from available literature that the proximity of wetlands to a water body may be more important to MeHg supply than simply the percent cover of wetlands in a given catchment. Likewise, Kramar et al. (this issue) determined that it was not the total area of wetlands, but rather the distribution of wetlands relative to loon nesting areas that controlled Hg concentrations in loons. Similarly, Branfireun et al. (1996) identified nearshore peatlands as important sources of MeHg to a downstream pond, whereas Bishop et al. (1995a) concluded that the near-stream riparian zone was the main source of MeHg to a small stream in Sweden.

Methylation efficiency

We also analyzed the controls on methylation efficiency, or MeHg/HgT. A multiple regression model dominated by stream discharge and the seasonal term explained 78% of the variance in MeHg/HgT ratios for the 79 cases that had both MeHg and discharge values. The discharge term was significant ($p = 0.004$) but its coefficient was negative, probably because as flow increases, HgT increases faster than MeHg increases. If discharge was removed as a variable, the number of cases in the model increased to 165 but the variance explained dropped to 32%, probably a consequence of broadening the data set to other study areas with a more diverse set of controls on both HgT and MeHg.

Summary of Hg in streams

Recapitulating the stream model results, watershed characteristics and climatic variables alone explained only 19% of the variance in HgT, and the inclusion of stream discharge as a predictor doubled the explanatory power of the model to 38%. For MeHg, watershed characteristics and climatic variables alone explained 25% of the variance and the inclusion of stream discharge increased predictive power more modestly to 33%. All of these percentages were rather low, likely the result of several factors: (1) the diverse nature of the study areas; (2) inter-study differences in methodologies; (3) the many missing data that reduced the number of cases analyzed; and (4) the lack of Hg deposition as an explanatory variable. Moreover, the respective models with and without stream discharge showed very different groupings and importance rankings of explanatory variables (Table 3), probably because the low-flow cases were not a true subset of the all-stream data set as discussed previously. Thus the actual importance of stream discharge to HgT and MeHg concentrations may be greater or less than indicated by the improvements in model performance. Dennis et al. (this issue) modeled HgT concentrations in surface waters across the landscape, and observed high HgT concentrations near some urbanized areas but also in remote, forested areas. Thus, multiple watershed characteristics may affect HgT concentrations. Our models suggest that concentration of MeHg is controlled by watershed characteristics to a greater degree than is HgT, and that conversely, HgT is more affected than is MeHg by the dynamics of high flow episodes.

Mercury in lakes

No single physical parameter emerged as a significant predictor of HgT concentrations in lakes (Table 3). For MeHg, a marginally significant model was constructed that explained only 9% of the variance in MeHg concentrations. The leading predictor in the lake MeHg model was percent wetlands, which in a single regression explained 7% of the variance ($p < 0.0001$) for the 202 cases included (Fig. 3). Analyzing essentially this same data set, Kamman et al. (2003) likewise found no statistical correlation between epilimnetic MeHg concentration and watershed percent wetlands, attributing the lack of correlation to the larger degree of heterogeneity in the types of lakes sampled. Subsequently, Kamman et al. (2004) applied principal components analysis to show that MeHg concentrations tended to be highest in lakes that were acidic or eutrophic. The limited ability of physical models of watershed characteristics to explain HgT and MeHg concentrations is likely because in-lake processes dominate over watershed processes in controlling lake HgT and MeHg concentrations (Watras et al., 1995).

Mercury in fish

On a set of 154 water bodies in New England and the Adirondacks, including 134 lakes and 20 reservoirs, we assessed physical factors controlling fish Hg concentrations. This set of lakes overlapped partially with those analyzed in the previous section. Hg concentrations in whole yellow perch were length-adjusted and expressed as deviation from the mean, as described in Kamman et al. (this issue). This resulting Hg concentration index was regressed against the same set of physical watershed characteristics (Table 4) as used in the aqueous Hg analyses.

For the 134 lakes, the best regression model explained 18% of the variance in fish Hg concentrations. The most significant predictor was percent wetlands ($p = 0.0002$) followed by percent forest ($p = 0.001$) and mean annual temperature ($p = 0.008$). All three parameters had positive correlations with fish Hg. Increasing percent wetlands and temperature promote methylation, so the positive correlation is logical. Percent forest may be an important predictor because of the tendency of the forest to enhance deposition.

For the 20 reservoirs, the best model explained 55% of the variance in fish Hg concentrations. Percent open water, mean soil permeability, and percent developed land were the three best predictors and were of approximately equal significance ($p \sim 0.005$). All three of these parameters were negatively correlated with fish Hg. One can argue that each of these parameters deters methylation, but conclusions are somewhat speculative given this small sample size.

The explanatory capability of these regression models suggests that watershed characteristics significantly affect fish tissue Hg concentrations in lakes and reservoirs. This finding stands in contrast to the poor ability of watershed characteristics to explain HgT and MeHg concentrations in lake waters, as discussed previously. Moreover, within this fish data set, watershed characteristics were more successful at predicting Hg concentration in fish than were HgT and MeHg concentrations in water and sediment measured directly in the lakes. Although Hg in fish usually does correlate to Hg in the water column and sediment (Sorensen et al., 1990; Kannan et al., 1998), in a subset of 45 lakes in this data set, Kamman et al. (2004) found no relation between Hg concentrations in yellow perch and Hg and MeHg concentrations in hypolimnetic lake water and sediments.

Conclusions

Across the diverse landscapes of the northeastern USA, total mercury concentrations in surface waters could be partially explained (<40%) by a broad set of physical variables. In streams under

Table 4. Results of multiple linear regression models for mercury concentrations in fish (length-normalized yellow perch) in lakes and reservoirs of northeastern USA in two categories

Category	*n*	Model r^2	Predictor	*p*-value
Lakes	134	0.183	Percent wetland	0.0002
			Percent forest	0.001
			Annual temperature	0.008
Reservoirs	20	0.546	*Percent open water*	0.003
			Soil permeability	0.006
			Percent urban	0.006

Predictors accepted in models only if $p < 0.25$. Predictors in italics have negative correlation. n = number of cases in each model.

low-flow conditions, static watershed factors were the best predictors, including percent forest cover for HgT concentrations (negative correlation) and percent wetland for MeHg. When stream discharge was included as a variable, it became the dominant predictor for HgT, doubling the explanatory power of the model from 19 to 38%. Stream discharge provided only modest additional explanatory power for MeHg concentrations in streams (from 25 to 33%). MeHg also had a strong seasonal component driven by temperature, which was reflected in a summer peak in the methylation efficiency (MeHg/HgT). The physical parameters had little success in predicting HgT and MeHg concentrations in lakes; only the MeHg model was marginally significant ($r^2 < 0.10$). Perhaps in-lake factors outweighed watershed physical variables in importance. Despite this lack of success in predicting aqueous Hg concentrations in lakes, we could predict 18% of the variance in Hg in yellow perch in 134 lakes with a model that had percent wetlands as its most significant factor.

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